Supporting Information

Boosting the lithium storage performance of

Na$_2$Li$_2$Ti$_6$O$_{14}$ anode by g-C$_3$N$_4$ modification

Ying Li [a,b], Fanfan Wang [a,b], Xue-Zhong Li [a,b], Xuan Gui [c],

Yan-Rong Zhu [b], Ping Cui [c,*], Ting-Feng Yi [a,b,c,d,*]

a School of Materials Science and Engineering, Northeastern University, Shenyang 110819, China

b School of Resources and Materials, Northeastern University at Qinhuangdao, Qinhuangdao 066004, China

c School of Chemistry and Chemical Engineering, Anhui University of Technology, Maanshan, Anhui 243002, China

d Key Laboratory of Dielectric and Electrolyte Functional Material Hebei Province, Qinhuangdao, China

1. Preparation of Na$_2$Li$_2$Ti$_6$O$_{14}$ and g-C$_3$N$_4$ coated Na$_2$Li$_2$Ti$_6$O$_{14}$

Na$_2$Li$_2$Ti$_6$O$_{14}$ was synthesized by a solid-state reaction. Primarily, 0.4076 g Li$_2$CO$_3$, 2.5176 g TiO$_2$ and 0.8619 g CH$_3$COONa were mixed. Then, the mixture was ground with ethanol in a planetary ball mill for 24 h and then dried at 70 °C for about 10 h. After that, the precursor was pre-treated at 400 °C for 4 h and then calcined at 800 °C for another 10 h in the air. After cooling down to room temperature naturally, the resulting product was ground in an agate mortar to form pristine Na$_2$Li$_2$Ti$_6$O$_{14}$.

The g-C$_3$N$_4$ (2.5, 5 and 8 wt%) coated Na$_2$Li$_2$Ti$_6$O$_{14}$ was also prepared by a facile solution method. The g-C$_3$N$_4$ was obtained by loading 6 g of melamine into a porcelain

boat and heated at 550 °C for 3 h. After natural cooling to room temperature, the yellow solid products were collected and grounded into fine g-C_3N_4 powder. A certain amount of $Na_2Li_2Ti_6O_{14}$ and g-C_3N_4 were mixed in absolute ethyl alcohol, and then ground 15 h. The obtained slurry was dried at 80 °C for 24 h in a vacuum oven, and then the g-C_3N_4 (2.5, 5 and 8 wt%) coated $Na_2Li_2Ti_6O_{14}$ powders were obtained.

2. Material characterization

The crystalline structure of all samples was measured by X-ray diffraction (XRD). The morphology and lattice structure of the samples were tested by a JSM-7800F Prime field emission scanning electron microscope (SEM) and high-resolution transmission electron microscope (HRTEM, JEOL, JEM-2010). Fourier transform infrared (FT-IR) spectroscopy of the samples was performed using a Nicolet Nexus 360 FT-IR spectrophotometer with a resolution of 4 cm–1. X-ray photoelectron spectroscopy (XPS, Escalab 250Xi, Thermo Fisher Scientific) with a Al Ka radiation was employed to observe the chemical states of GNLTO-2. The energy-dispersive spectroscopic spectrum (EDS) element mapping was performed using a Bruker-QUANTAX instrument.

3. Electrochemical characterization

The homogeneous slurry containing active material, acetylene black, polyvinylidene fluoride (PVDF) (80:10:10 in weight) and N-methyl-pyrrolidone (NMP) were painted on a copper foil and desiccated at 110 °C for 13 h under vacuum to form the working electrode. CR2025 coin-type cells were employed to investigate the electrochemical performance of all samples. The half-cell consists of working electrode and lithium foil,

which separated by a porous polypropylene film, and 1 mol L^{-1} $LiPF_6$ in EC: DMC (1:1 in volume) as the electrolyte. The loading mass of active material is about 2.5 mg cm^{-2}. The cyclic voltammetry (CV) test was performed on a CHI 1000C (Shanghai, China) electrochemical workstation at a scanning rate of 0.2 mV s^{-1} between 0.01 and 3 V. Electrochemical impedance spectroscopy (EIS) studies were carried out on a Princeton P4000 (American) electrochemical workstation with a frequency of 100 kHz to 0.01 Hz. The galvanostatic charge-discharge performance were conducted between 0.01 and 3 V (*vs.* Li^+/Li) on LANHE CT2001A (Wuhan, China).

4. **Nomenclature for equations (1) and (2)**

A the surface area of the electrode

T the absolute temperature

R the gas constant

F the Faraday constant

n the number of electrons per molecule during oxidation
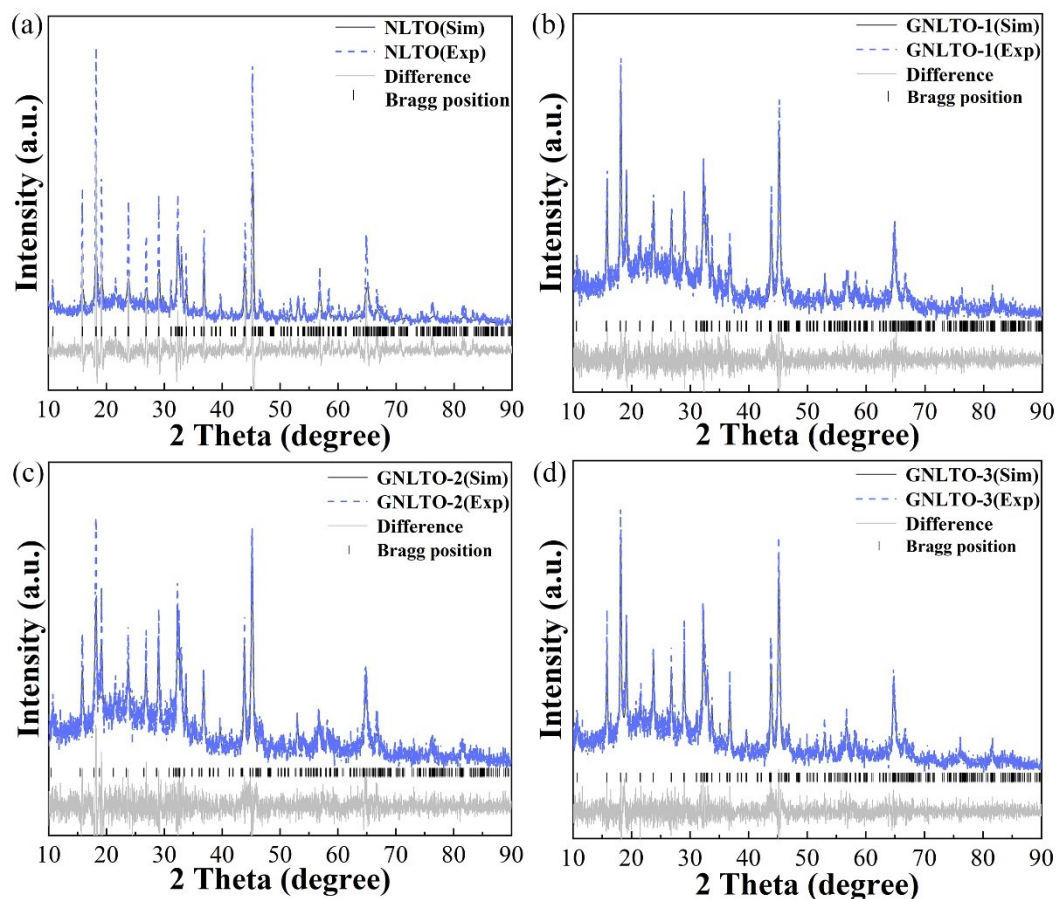
C_{Li} the concentration of lithium ion

Fig. S1 XRD refinement plots of pristine $Na_2Li_2Ti_6O_{14}$ and $Na_2Li_2Ti_6O_{14}@g-C_3N_4$ composite materials (a)NLTO, (b) GNLTO-1, (c) GNLTO-2 and (d) GNLTO-3
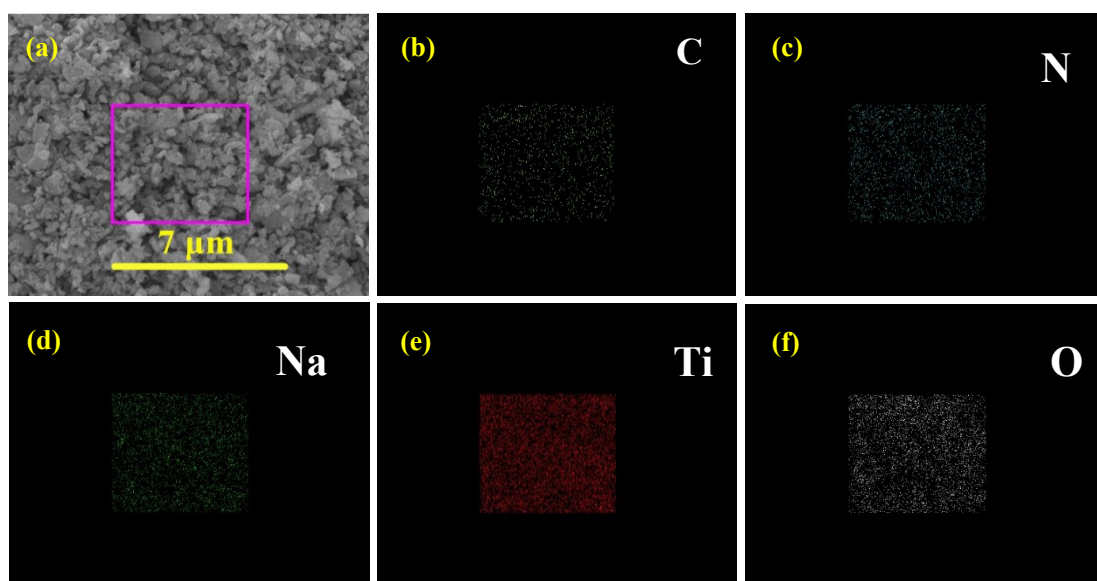

Fig. S2 (a) SEM, (b-f) EDS mapping images of GNLTO-2 composite material